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Cation Ordering in $Pb(Sc_{2/3}W_{1/3})O_3$ - $PbMO_3$, M = Ti, Zr relaxors

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Beamline(s): X7A

Introduction: Lead-containing perovskites are widely utilized as capacitors and transducers. For the $PbBO_3$ systems containing a mixture of cations at the octahedral B site, the degree of chemical order has been found to be critical for the overall dielectric behavior [1]. This study focuses on materials based on $Pb(Sc_{2/3}W_{1/3})O_3$ (PSW), which forms a B-site ordered structure with a 1:1 type periodicity. In our previous work, the effects of B-cation substitutions and thermal treatment on the B-site order have been examined for two solid solutions, (1-*x*)PSW - (*x*)$PbTiO_3$ and (1-*x*)PSW - (*x*)$PbZrO_3$. For PSW-PT the B-site order was found to be much more stable, extending to higher substitution levels than in the PSW-PZ system. All of the investigated compositions showed relaxor ferroelectric behavior, however PSW-PT displayed unusual trends in the temperature of the permittivity maximum T_{max} which decreased for $x \leq 0.2$ and then increased for $x > 0.25$ - Fig. 1. In contrast, for the PZ system T_{max} varies linearly with the Zr content - Fig. 2. These observations can be rationalized in terms of the B-occupancies predicted by the "random-site" model. According to this model, the structure of the fully ordered PSW end-member can be represented as $Pb[Sc]_{1/2}[Sc_{1/3}W_{2/3}]_{1/2}O_3$ and the ordered solid solutions by $Pb[Sc]_{1/2}[Sc_{(1-4x)/3}W_{(2-2x)/3}M_{2x}]_{1/2}O_3$, where M^{4+} is either Zr or Ti. In order to confirm this model, an accurate structure analysis is required to quantify the B-site occupancies. Temperature factors obtained by such analysis should indicate the magnitude of Pb and O displacements, which also play a critical role in mediating the dielectric behavior [2].

Methods and Materials: Samples of PSW-PZ and PSW-PT, $x \leq 0.35$ were prepared by solid-state methods from high-purity oxides (>99.9%) via the "columbite route". X-ray diffraction data were collected at NSLS X7A beamline using the PSD detector. A Si (111) crystal was used as a monochromator for the incident beam and the wavelength of (0.69992Å) was calibrated using a Si standard. Powder specimens were placed in a thin glass capillary, which was rotated during the experiment. The data were analyzed by a Rietveld method using a GSAS 2000 software package.

Results: All of the investigated compounds showed cubic symmetry with no sign of tetragonal or orthorombic phases. Compositions of PSW-PT, $x \leq 0.35$ and PSW-PZ, $x \leq 0.15$ exhibited 1:1 B-site order with a doubled unit cell, S.G. *Fm-3m*. No B-site order has been detected for PSW-PZ, $x = 0.35$, suggesting a random arrangements of B-cations, S. G. *Pm-3m*. The structure analysis is still in progress, however preliminary values for B-occupancies are in a good agreement with the random site model.

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References:

[1] P. K. Davies, M. A. Akbas, J. of Phys. and Chem. of Solids, 61 (2): 159-166, 2000

[2] W. Dmowski, M. A. Akbas, P. K. Davies, et al., J. of Phys. and Chem. of Solids, 61 (2): 229-237, 2000

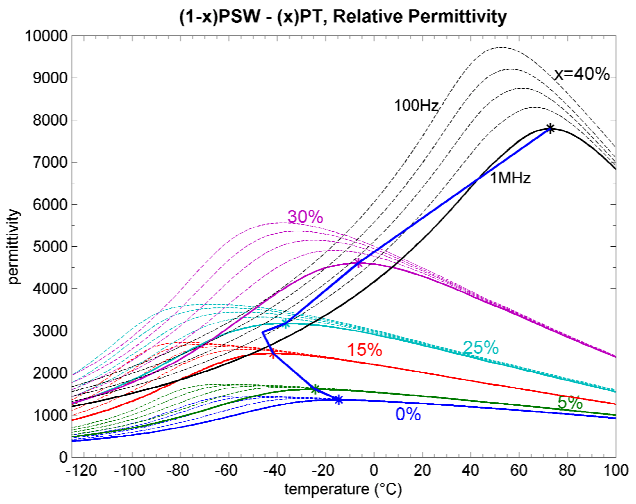

Figure 1: - Permittivity for PSW-PT

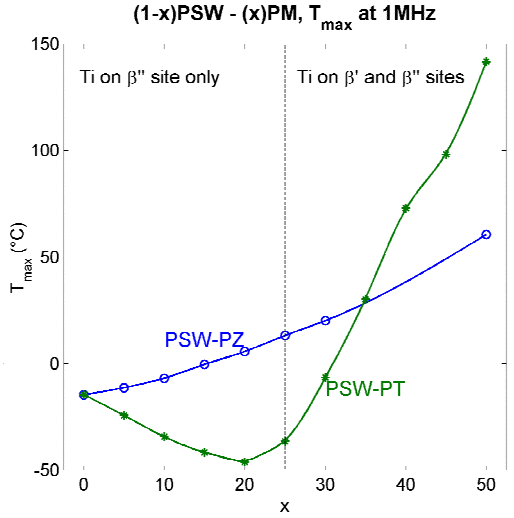

Figure 2: - Temperature of permittivity maximum in PSW-PT and PSW-PZ at 1 MHz